Response to Comments on the Manuscript (acp-2020-1004):

"Estimating daily full-coverage and high-accuracy 5-km ambient particulate matters across China: considering their precursors and chemical compositions"

Response to Comments of Referee #2:

General comment:

In this paper, Wang et al. proposed a framework to estimate the daily PM2.5 and PM10 concentration over China by combining multiple data sources with a Light Gradient Boosting Machine learning method. They included satellite product (TROPOMI) and modeled data assimilation dataset (GEOS-FP) as main predictors. Even though they showed some reasonable statistics from the validation process, the advantages of this method are not well justified. Also, some flaws are found in their validation process. As a result, I do not recommend this paper for publication in ACP.

Response: We would like to express our sincere gratitude to the referee for his/her comments and recommendations for improving the paper. An item-by-item response to the comments raised by the referee follows. Thanks for your time.

Major comments:

Q2.1: The selection of predicting variables from TROPOMI is too arbitrary and lacks justification. In this paper, the authors only considered column NOx and SO2 observations, which are the precursors of sulfate and nitrate. Both of them are large components of PM2.5. However, other components are also important. For example organic aerosols. Why the precursors of organic aerosols were not chosen as input predictors? Also, this idea of "PM is

associated with ozone, so choose column ozone as one of the predictors" needs more justifications.

Response: Thank the referee for his/her significant comments. The purpose of our study is to estimate daily full-coverage PM at high spatial resolution using the datasets of their precursors & chemical compositions instead of AOD products. Therefore, the selection of predicting variables should be carefully considered. As for remote sensing sensors, only TROPOMI can generate daily high-spatial-resolution (e.g., 5-km) and high-coverage chemical species at present. In our study, the atmospheric products of NO_2 and SO_2 from TROPOMI were adopted, regarded as two precursors of PM. However, there is merely one precursor of organic aerosol, i.e., formaldehyde, belonging to the atmospheric products from TROPOMI (see https://earth.esa.int/web/guest/missions/esa-eo-missions/sentinel-5p). Since formaldehyde is generally not a major organic aerosol precursor (Hallquist et al., 2009; Volkamer et al., 2006), this product was not utilized. By contrast, the Organic Carbon Column Mass Density (carbon-related) from GEOS-FP was used in this paper. Furthermore, some other chemical compositions of PM were also acquired from GEOS-FP, including the Nitrate Column Mass Density (nitrate-related), SO4 Column Mass Density (sulfate-related), Black Carbon Column Mass Density (carbon-related), Dust Column Mass Density (dust-related), Ammonium Column Mass Density (ammonium-related), and Sea Salt Column Mass Density (sea salt-related). All of these variables are employed according to the major chemical compositions of PM (i.e., nitrate, sulfate, carbon, dust, ammonium, and sea salt) (Baker et al., 2007; Tucker et al., 2000; Zheng et al., 2005; Wang et al., 2019; Pui et al., 2014). We have appended this statement in the manuscript. It is concluded that the selected variables are sufficient to estimate PM over China. The validation results show that the estimation model achieves a satisfactory performance (e.g., space-based CV R^2: 0.88 for $PM_{2.5}$ and 0.83 for PM_{10}) in our study, which also signify this point.

The justification for the adoption of total O_3 column is presented as follows. With regard to stratospheric O_3, a latest study (Chen et al., 2020) has shown that the downward transport of O_3 stemming from the stratosphere-to-troposphere exchange can be a significant contributor to background O_3. Such enhancement of background O_3 will affect ambient PM. In addition, ambient O_3 pollution is rapidly increasing over China in recent years (Liu et al., 2020; Wang et al., 2020) and the proportion of it may also rise in the total O_3 column. At present, the total O_3 column has been used to estimate ambient O_3 over China (Liu et al., 2020) and Tibetan Plateau (Li et al., 2020), suggesting its surface predictive capacity. In China, ambient PM is associated with ambient O_3 (Chen et al., 2019). Therefore, the total O_3 column is introduced as an auxiliary variable in our study.

The main revision is as follows:

In our study, all of the chemical species are selected in accordance with the major component of $PM_{2.5}$ and PM_{10} (i.e., nitrate, sulfate, carbon, dust, ammonium, and sea salt) (Baker et al., 2007; Tucker et al., 2000; Zheng et al., 2005; Wang et al., 2019; Pui et al., 2014).

References:

Baker, K. and Scheff, P.: Photochemical model performance for PM2.5 sulfate, nitrate, ammonium, and precursor species SO2, HNO3, and NH3 at background monitor locations in the central and eastern United States, Atmos. Environ., 41(29), 6185-6195, 2007.

Chen, L., Xing, J., Mathur, R., Liu, S., Wang, S., & Hao, J. (2020). Quantification of the enhancement of PM2.5 concentration by the downward transport of ozone from the stratosphere. Chemosphere, 126907.

Chen, J., Shen, H., Li, T., Peng, X., Cheng, H., & Ma, C. (2019). Temporal and Spatial Features of the Correlation between PM2.5 and O3 Concentrations in China. International Journal of Environmental Research and Public Health, 16(23), 4824.

Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., ... & Wildt, J. (2009). The formation, properties and impact of secondary organic aerosol: current and emerging issues. Atmospheric chemistry and physics, 9(14), 5155-5236.

Li, R., Zhao, Y., Zhou, W., Meng, Y., Zhang, Z., & Fu, H. (2020). Developing a novel hybrid model for the estimation of surface 8 h ozone (O3) across the remote Tibetan Plateau during 2005–2018. Atmospheric Chemistry and Physics, 20(10), 6159-6175.

Liu, R., Ma, Z., Liu, Y., Shao, Y., Zhao, W., & Bi, J. (2020). Spatiotemporal distributions of surface ozone levels in China from 2005 to 2017: A machine learning approach. Environment International, 142, 105823.

Pui, D. Y., Chen, S. C., & Zuo, Z. (2014). PM2.5 in China: Measurements, sources, visibility and health effects, and mitigation. Particuology, 13, 1-26.

Tucker, W. G.: An overview of PM2.5 sources and control strategies, Fuel Process. Technol., 65, 379-392, 2000.

Volkamer, R., Jimenez, J. L., San Martini, F., Dzepina, K., Zhang, Q., Salcedo, D., ... & Molina, M. J. (2006). Secondary organic aerosol formation from anthropogenic air pollution: Rapid and higher than expected. Geophysical Research Letters, 33(17).

Wang, Y., Wild, O., Chen, X., Wu, Q., Gao, M., Chen, H., ... & Wang, Z. (2020). Health impacts of long-term ozone exposure in China over 2013–2017. Environment International, 144, 106030.

Wang, Y., Li, W., Gao, W., Liu, Z., Tian, S., Shen, R., and Song, T.: Trends in particulate matter and its chemical compositions in China from 2013–2017, Science China Earth Sciences., 62(12), 1857-1871, 2019.

Zheng, M., Salmon, L. G., Schauer, J. J., Zeng, L., Kiang, C. S., Zhang, Y., & Cass, G. R. (2005). Seasonal trends in PM2. 5 source contributions in Beijing, China. Atmospheric Environment, 39(22), 3967-3976.

Q2.2: By using GEOS-FP, this method loses its flexibility to adjust its input conditions. GEOS-FP is one of the data assimilation products from GMAO. It is based largely on the model output of GEOS. The method in this paper is largely based on this dataset. According to their fig 6, 4 out of 6 top feature importance for their PM2.5 prediction are from GEOS-FP. What this means is that their prediction is mostly controlled by a dataset that they could not make any modifications to. For example, the GEOS-FP system has been updated to version 5.25 in January 2020. Let alone some discontinuous issues posed by all the updates along with the release of each GEOS-FP products. On the other hand, the authors argue that previous studies using CTMs have the limitation of large uncertainty in the emission inventories of the CTMs. By saying that, GEOS-FP also has the same problem of large uncertainty in their emission inventories. By using CTMs instead of data assimilation datasets as prediction inputs, researchers can do their best to narrow the uncertainty in their model simulations and could also conduct sensitivity experiments. From this point of view, methods using CTMs or earth models (e.g. GEOS) would be better for this kind of prediction.

Response: Thank the referee for his/her valuable comments. To be clearer, the total comments are divided into two parts, which are replied as follows.

"By using GEOS-FP, this method loses its flexibility to adjust its input conditions. GEOS-FP is one of the data assimilation products from GMAO. It is based largely on the model output of GEOS. The method in this paper is largely based on this dataset. According to their fig 6, 4 out of 6 top feature importance for their PM2.5 prediction are from GEOS-FP. What this means is that their prediction is mostly controlled by a dataset that they could not make any modifications to. For example, the GEOS-FP system has been updated to version 5.25 in January 2020. Let alone some discontinuous issues posed by all the updates along with the release of each GEOS-FP products."

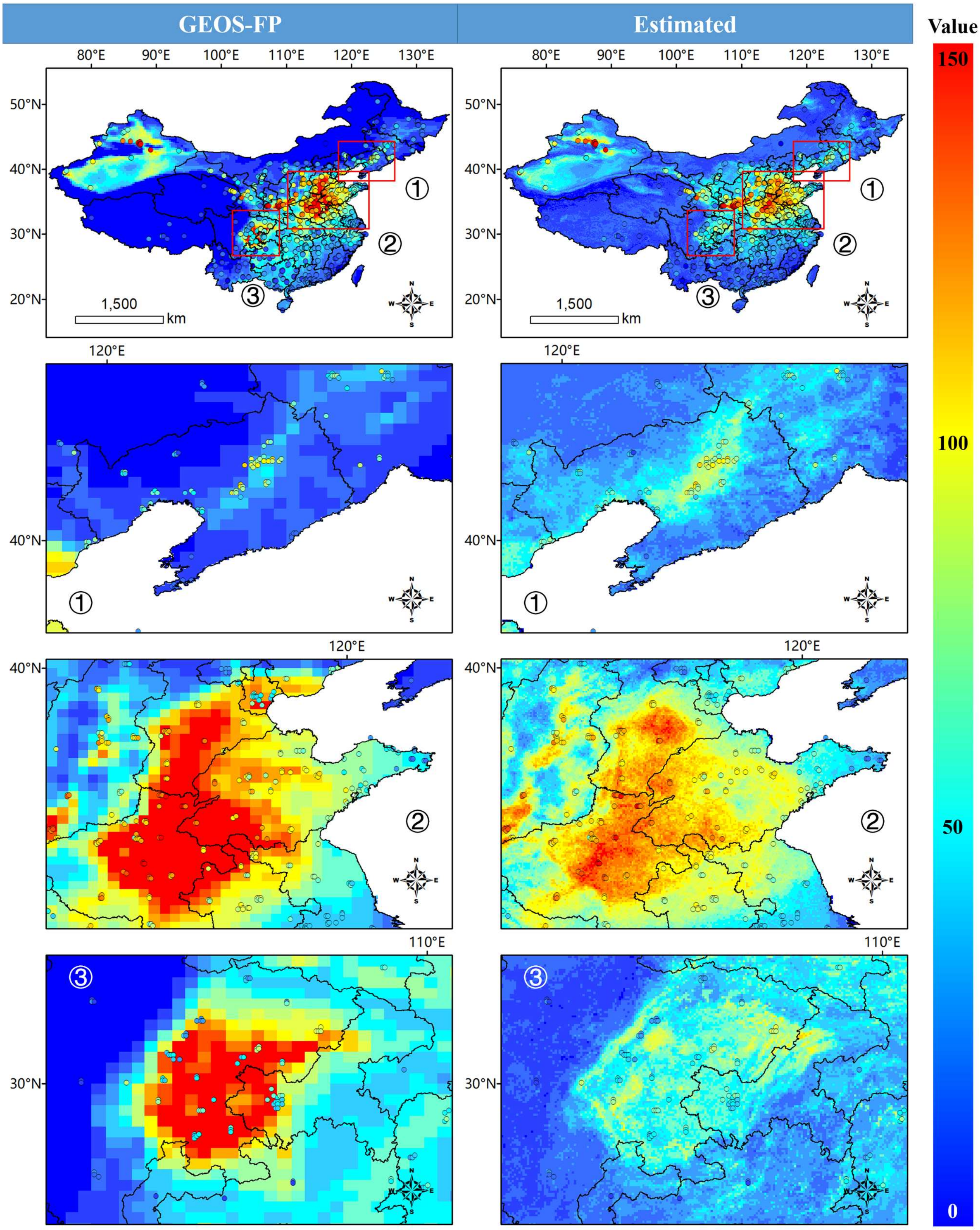

Figure r1： Daily (20190101) spatial distribution of the GEOS-FP, estimated, and ground-based $PM_{2.5}$. The circles represent the ground-based sites.

Apart from GEOS-FP, multiple datasets from other sources were adopted in our study, including

TROPOMI, MODIS, GPW, and OpenStreetMap. Especially, high-resolution geographical factors, such as NDVI, road density, and population density, could maintain the spatial information. In addition, an advance ensemble learning method, i.e., light gradient boosting machine, was exploited to fuse the multisource data using ground-truth values. Space-based CV results show that the proposed framework performs well in the study area (e.g., R^2: 0.88 for $PM_{2.5}$ and 0.83 for PM_{10}), suggesting that GEOS-FP data did not introduce large uncertainties. An example to compare the spatial distribution between the estimated and GEOS-FP $PM_{2.5}$ is demonstrated in Figure r1. The GEOS-FP $PM_{2.5}$ is calculated via this formula: $PM_{2.5}$=1.375*SO_4+2.1*OC+BC+$DS_{2.5}$+$SS_{2.5}$ (Xiao et al., 2018). As can be seen, the spatial patterns of the estimated $PM_{2.5}$ are much closer to the actual measurements than GEOS-FP, with a higher spatial resolution. Therefore, the estimation accuracy is greatly improved compared to GEOS-FP based on posterior techniques.

"On the other hand, the authors argue that previous studies using CTMs have the limitation of large uncertainty in the emission inventories of the CTMs. By saying that, GEOS-FP also has the same problem of large uncertainty in their emission inventories. By using CTMs instead of data assimilation datasets as prediction inputs, researchers can do their best to narrow the uncertainty in their model simulations and could also conduct sensitivity experiments. From this point of view, methods using CTMs or earth models (e.g. GEOS) would be better for this kind of prediction."

In this paper, the previous studies using CTMs refer to the works merely considering CTMs without other techniques. Our study does not focus on comparing to or arguing about the methods using CTMs. The mention of CTMs in the introduction was only to elicit that the approaches based on remote sensing satellites have been greatly developed in recent years. The statements about CTMs is confusing and has been rephrased in the manuscript. CTMs and GEOS-FP both potential present large uncertainty in their emission inventories. At present, the outputs of CTMs have been combined with other datasets (e.g., remote sensing) to estimate $PM_{2.5}$, such as the mentioned work (van Donkelaar et al., 2019). In our study, we also fused multiple datasets from new sources, including new remote sensing sensor (TROPOMI) and new data assimilation (GEOS-FP). Moreover, the present study is a novel attempt to estimate daily full-coverage PM based on the datasets of their precursors & chemical compositions instead of AOD products. The validation results show that the proposed framework can perform well without the input of AOD in the study area.

The main revision is as follows:

With regard to CTMs, the uncertainties of the emission inventories could be large in some areas (Li et al., 2017b) and it will consume time and energy to collect the necessary information for simulation (Chu et al., 2016). The approaches based on remote sensing satellites have been greatly developed in recent years (Sorek-Hamer et al., 2020).

References:

Xiao, Q., Chang, H. H., Geng, G., & Liu, Y. (2018). An ensemble machine-learning model to predict historical PM2. 5 concentrations in China from satellite data. Environmental science & technology, 52(22), 13260-13269.

van Donkelaar, A., Martin, R. V., Li, C., & Burnett, R. T. (2019). Regional estimates of chemical composition of fine particulate matter using a combined geoscience-statistical method with information from satellites, models, and monitors. Environmental science & technology, 53(5), 2595-2611.

Q2.3: The validation process exists flows. First, the authors defined their "AOD-based" control estimate, which is using VIIRS AOD to replace the TROPOMI and GEOS-FP in their estimating framework. However, this is not the case of previous AOD based studies, especially those most influential ones, for example, van Donkelaar et al. 2019. Previous AOD based studies usually combined with model simulations and ground-based measurements to best use the information of satellite AOD products. What has been done in the paper was that comparing an estimate from a missing data AOD product with an estimate from the combination of satellite observations and model output. So that it caused the issue of comparing apple with orange; moreover, the selling point of this paper is the daily PM2.5 estimate. So validation regarded to a daily resolution would be the most convincing. Otherwise, why not just estimate the seasonal or annual PM concentrations, which are more useful in current epidemiological studies. The validation results from the time-based cross-validation method were the worst according to their table1.

Response: Thank the referee for his/her important comments. To fully validate the estimated results, a total of three CV schemes (i.e., sample-based, space-based, and time-based) were considered in our study. Meanwhile, all of the validation results were performed at daily resolution, including the overall validation (Figure 4), regional validation (Table 1), seasonal validation (Fig. S3-S6), and grid-based validation (Figure 5, S7-S9). The examples of daily estimated results in different seasons (20190122, 20190501, 20190803, and 20191111) were also provided in Figure 7 to show the daily spatial distributions.

Moreover, a baseline was devised in our study. Since a large number of related studies over China based on machine learning methods only adopted remote sensing AOD products (no model simulations) to estimate PM (Li et al., 2020; Yang et al., 2019; Wei et al., 2019; He et al., 2018, 2020; Yao et al., 2019; Ma et al., 2016; Chen et al., 2018a, 2018b, 2019; Wang et al., 2019; Zhang et al., 2019; Xue et al., 2020), the VIIRS DB AOD product was selected as the AOD-based. However, related studies could utilize various techniques to improve their estimation performance. For instance, Li et al. (2020) proposed GTWNN; Wei et al. (2019) developed STRF; Kong et al. (2020) combined model simulations and ground-based measurements. It is very difficult to duplicate their experiments as baselines due to different hardware facilities, large time consumptions, closed data sources, unspecified model parameters, etc. By contrast, we compared our metrics to those of related studies over China in recent years, which are listed in Table r1 (or see Table S5 in the supplementary materials). This is a common strategy used in previous studies for comparison with other works (Wei et al., 2019; Jiang et al., 2020; Kong et al., 2020). Apart from Kong et al. (2020) (model simulations), another two related studies over China using model simulations or reanalysis datasets have been appended in the table (Xue et al., 2017; Xiao et al., 2018), as the referee pointed out. The study area of the mentioned work (van Donkelaar et al., 2019) is North America and its temporal resolution is annual. Therefore, this work cannot be compared in our study.

Table r1: Detailed information about the previous related works over China. SACV: sample-based CV; SPCV: space-based CV; TICV: time-based CV; SR: spatial resolution; TR: temporal resolution; FC: full-coverage; T: true; F: false; MF: the factors which lead to the missing values in the estimated results.

Type	Reference	Metric	SACV	SPCV	TICV	SR	TR	Study period	FC	MF
	Proposed	R^2	0.93	0.88	0.73	5-km	Daily	2019※	T	None
		RMSE	8.87 µg/m^3	11.56 µg/m^3	17.3 µg/m^3					
		RPE	22.8%	29.8%	44.5%					
$PM_{2.5}$	Wei et al., 2019	R^2	0.85	0.83	0.63	1-km	Daily	2016	F	Cloud, snow/ice
		RMSE	15.57 µg/m^3	16.63 µg/m^3	24.83 µg/m^3					
		RPE	-	-	-					
	He et al., 2018	R^2	0.8	-	-	3-km	Daily	2015	F	Cloud, snow/ice, bright surface
		RMSE	18 µg/m^3							

		RPE	-							
	Yao et al., 2019	R^2		0.6		6-km	Daily	2014	F	Cloud, snow/ice, bright surface
		RMSE	-	21.76 $\mu g/m^3$	-					
		RPE		-						
	Li et al., 2020	R^2	0.8	0.79		10-km	Daily	2015	F	Cloud, snow/ice
		RMSE	17.38 $\mu g/m^3$	17.81 $\mu g/m^3$	-					
		RPE	31.5%	32.29%						
	Jiang et al., 2020	R^2	0.85	0.74		1-km	Daily*	2018.03.01-2019.02.28	T	None
		RMSE	11.02 $\mu g/m^3$	14.65 $\mu g/m^3$	-					
		RPE	-	-						
	Kong et al., 2020	R^2		0.86		15-km	Daily*	2013–2018	T	None
		RMSE	-	15.1 $\mu g/m^3$	-					
		RPE		-						
	Xue et al., 2017	R^2		0.72		10-km	Daily	2014	T	None
		RMSE	-	23 $\mu g/m^3$	-					
		RPE		41%						
	Xiao et al., 2018	R^2	0.79	0.76	0.73	10-km	Daily	2013-2016	T	None
		RMSE	-	-	-					
		RPE								
PM_{10}	Proposed	R^2	0.91	0.84	0.67	5-km	Daily	2019※	T	None
		RMSE	16.92 $\mu g/m^3$	22.03 $\mu g/m^3$	31.33 $\mu g/m^3$					
		RPE	24.5%	31.9%	45.4%					
	Chen et al., 2018b	R^2		0.78		10-km	Daily	2005–2016	F	Cloud, snow/ice
		RMSE	-	31.54 $\mu g/m^3$	-					
		RPE		-						
	Kong et al., 2020	R^2		0.81		15-km	Daily*	2013–2018	T	None
		RMSE	-	28.8 $\mu g/m^3$	-					
		RPE		-						

Note:
1. The symbols of * represent that the works could provide the estimated results at various temporal resolutions, while the metrics listed in the table are computed from the daily estimation.
2. ※: Only the metrics computed from the estimated results through the proposed approach for a whole year (2019) are listed in the table to be fairly compared to previous works. The study period of this paper is from June 1, 2018 to March 31, 2020.

The validation results from the time-based CV were the worst since the temporal heterogeneity of PM is usually strong (Li et al., 2017, 2020). In other word, the temporal variations of PM could not be fully captured. This phenomenon can be discovered in previous related studies over China, such as

Wei et al. (2019) (time-based CV R^2: 0.63) and Xiao et al. (2018) (time-based CV R^2: 0.73). Compared to them, the time-based CV results in our study are acceptable.

In conclusion, it is believed that the validation process was justified in this paper.

References:

Chen, G., Li, S., Knibbs, L. D., Hamm, N. A., Cao, W., Li, T., ... & Guo, Y. (2018a). A machine learning method to estimate PM2. 5 concentrations across China with remote sensing, meteorological and land use information. Science of the Total Environment, 636, 52-60.

Chen, J., Yin, J., Zang, L., Zhang, T., & Zhao, M. (2019). Stacking machine learning model for estimating hourly PM2. 5 in China based on Himawari 8 aerosol optical depth data. Science of The Total Environment, 697, 134021.

Chen, G., Wang, Y., Li, S., Cao, W., Ren, H., Knibbs, L. D., and Guo, Y. (2018b). Spatiotemporal patterns of PM10 concentrations over China during 2005–2016: A satellite-based estimation using the random forests approach, Environmental Pollution., 242, 605-613.

He, Q., Gu, Y., & Zhang, M. (2020). Spatiotemporal trends of PM2. 5 concentrations in central China from 2003 to 2018 based on MAIAC-derived high-resolution data. Environment international, 137, 105536.

He, Q. and Huang, B. (2018): Satellite-based mapping of daily high-resolution ground PM2.5 in China via space-time regression modeling, Remote Sensing of Environment., 206, 72-83.

Jiang, T., Chen, B., Nie, Z., Zhehao, R., Xu, B., and Tang, S. (2020). Estimation of hourly full-coverage PM2.5 concentrations at 1-km resolution in China using a two-stage random forest model, Atmospheric Research., 105146.

Kong, L., Tang, X., Zhu, J., Wang, Z., Li, J., Wu, H., and Liu, B.: A Six-year long (2013–2018) High-resolution Air Quality Reanalysis Dataset over China base on the assimilation of surface observations from CNEMC. (2020). Earth System Science Data Discussions., 1-44.

Li, T., Shen, H., Yuan, Q., Zhang, X., & Zhang, L. (2017). Estimating ground‐level PM2. 5 by fusing satellite and station observations: a geo‐intelligent deep learning approach. Geophysical Research Letters, 44(23), 11-985.

Li, T., Shen, H., Yuan, Q., and Zhang, L. (2020). Geographically and temporally weighted neural networks for satellite-based mapping of ground-level PM2.5, ISPRS Journal of Photogrammetry and Remote Sensing., 167, 178-188.

Ma, Z., Hu, X., Sayer, A. M., Levy, R., Zhang, Q., Xue, Y., ... & Liu, Y. (2016). Satellite-based spatiotemporal trends in PM2. 5 concentrations: China, 2004–2013. Environmental health perspectives, 124(2), 184-192.

Van Donkelaar, A., Martin, R. V., Li, C., & Burnett, R. T. (2019). Regional estimates of chemical composition of fine particulate matter using a combined geoscience-statistical method with information from satellites, models, and monitors. Environmental science & technology, 53(5), 2595-2611.

Wang, W., Zhao, S., Jiao, L., Taylor, M., Zhang, B., Xu, G., & Hou, H. (2019). Estimation of PM2. 5 concentrations in China using a spatial back propagation neural network. Scientific reports, 9(1), 1-10.

Wei, J., Huang, W., Li, Z., Xue, W., Peng, Y., Sun, L., & Cribb, M. (2019). Estimating 1-km-resolution PM2. 5 concentrations across China using the space-time random forest approach. Remote Sensing of Environment, 231, 111221.

Xue, Y., Li, Y., Guang, J., Tugui, A., She, L., Qin, K., ... & Wang, Z. (2020). Hourly PM2. 5 Estimation over Central and Eastern China Based on Himawari-8 Data. Remote Sensing, 12(5), 855.

Xue, T., Zheng, Y., Geng, G., Zheng, B., Jiang, X., Zhang, Q., & He, K. (2017). Fusing observational, satellite remote sensing and air quality model simulated data to estimate spatiotemporal variations of PM2. 5 exposure in China. Remote Sensing, 9(3), 221.

Xiao, Q., Chang, H. H., Geng, G., & Liu, Y. (2018). An ensemble machine-learning model to predict historical PM2.5 concentrations in China from satellite data. Environmental science & technology, 52(22), 13260-13269.

Yang, L., Xu, H., & Jin, Z. (2019). Estimating ground-level PM2. 5 over a coastal region of China using satellite AOD and a combined model. Journal of Cleaner Production, 227, 472-482.

Yao, F., Wu, J., Li, W., and Peng, J. (2019). A spatially structured adaptive two-stage model for retrieving ground-level PM2.5 concentrations from VIIRS AOD in China, ISPRS Journal of Photogrammetry and Remote Sensing., 151, 263-276.

Zhang, T., Zang, L., Wan, Y., Wang, W., & Zhang, Y. (2019). Ground-level PM2. 5 estimation over urban agglomerations in China with high spatiotemporal resolution based on Himawari-8. Science of the total environment, 676, 535-544.

Specific comments:

Q2.4: Line 99: is it necessary to mention that the area of this study was chosen because that China has the largest population in the world? Maybe rephrase this sentence or delete it.

Response: Thank the referee for his/her comment. This statement has been removed in the manuscript.

Q2.5: Line 106: "monitor"? maybe use "estimate".

Response: Thank the referee for his/her comment. The word "monitor" has been replaced with "estimate" in the manuscript.

The main revision is as follows:

It is necessary to develop an approach that can estimate $PM_{2.5}$ and PM_{10} across China continuously and precisely.

Q2.6: Line 120: any publications about the data validation, calibration, and uncertainty analysis of the Chinese PM2.5/PM10 measurements from CNEMC?

Response: Thank the referee for his/her comment. The required contents have been appended in the manuscript. The ground-based measurements from CNEMC have been widely used to estimate PM across China (Li et al., 2020; Yang et al., 2019; Wei et al., 2019; He et al., 2018, 2020; Yao et al., 2019; Ma et al., 2016; Chen et al., 2018a, 2018b, 2019; Wang et al., 2019; Zhang et al., 2019; Xue et al., 2017, 2019, 2020; Kong et al., 2020), suggestion their reliability and accuracy.

The main revision is as follows:

The CNEMC can provide hourly ambient concentrations of $PM_{2.5}$ and PM_{10} over China, which are obtained according to the technical specification of HJ 817-2018 (i.e., tapered element oscillating microbalance method or beta-attenuation method).

References:

Chen, G., Li, S., Knibbs, L. D., Hamm, N. A., Cao, W., Li, T., ... & Guo, Y. (2018a). A machine learning method to estimate PM2. 5 concentrations across China with remote sensing, meteorological and land use information. Science of the Total Environment, 636, 52-60.

Chen, J., Yin, J., Zang, L., Zhang, T., & Zhao, M. (2019). Stacking machine learning model for estimating hourly PM2. 5 in China based on Himawari 8 aerosol optical depth data. Science of The Total Environment, 697, 134021.

Chen, G., Wang, Y., Li, S., Cao, W., Ren, H., Knibbs, L. D., and Guo, Y. (2018b). Spatiotemporal patterns of PM10 concentrations over China during 2005–2016: A satellite-based estimation using the random forests approach, Environmental Pollution., 242, 605-613.

He, Q., Gu, Y., & Zhang, M. (2020). Spatiotemporal trends of PM2. 5 concentrations in central China from 2003 to 2018 based on MAIAC-derived high-resolution data. Environment international, 137, 105536.

He, Q. and Huang, B. (2018): Satellite-based mapping of daily high-resolution ground PM2.5 in China via space-time regression modeling, Remote Sensing of Environment., 206, 72-83.

Kong, L., Tang, X., Zhu, J., Wang, Z., Li, J., Wu, H., and Liu, B.: A Six-year long (2013–2018) High-resolution Air Quality Reanalysis Dataset over China base on the assimilation of surface observations from CNEMC. (2020). Earth System Science Data Discussions., 1-44.

Li, T., Shen, H., Yuan, Q., and Zhang, L. (2020). Geographically and temporally weighted neural networks for satellite-based mapping of ground-level PM2.5, ISPRS Journal of Photogrammetry and Remote Sensing., 167, 178-188.

Ma, Z., Hu, X., Sayer, A. M., Levy, R., Zhang, Q., Xue, Y., ... & Liu, Y. (2016). Satellite-based spatiotemporal trends in PM2. 5 concentrations: China, 2004–2013. Environmental health perspectives, 124(2), 184-192.

Wang, W., Zhao, S., Jiao, L., Taylor, M., Zhang, B., Xu, G., & Hou, H. (2019). Estimation of PM2. 5 concentrations in China using a spatial back propagation neural network. Scientific reports, 9(1), 1-10.

Wei, J., Huang, W., Li, Z., Xue, W., Peng, Y., Sun, L., & Cribb, M. (2019). Estimating 1-km-resolution PM2. 5 concentrations across China using the space-time random forest approach. Remote Sensing of Environment, 231, 111221.

Xue, T., Zheng, Y., Tong, D., Zheng, B., Li, X., Zhu, T., & Zhang, Q. (2019). Spatiotemporal continuous estimates of PM2. 5 concentrations in China, 2000–2016: A machine learning method with inputs from satellites, chemical transport model, and ground observations. Environment international, 123, 345-357.

Xue, Y., Li, Y., Guang, J., Tugui, A., She, L., Qin, K., ... & Wang, Z. (2020). Hourly PM2. 5 Estimation over Central and Eastern China Based on Himawari-8 Data. Remote Sensing, 12(5), 855.

Xue, T., Zheng, Y., Geng, G., Zheng, B., Jiang, X., Zhang, Q., & He, K. (2017). Fusing observational, satellite remote sensing and air quality model simulated data to estimate spatiotemporal variations of PM2. 5 exposure in China. Remote Sensing, 9(3), 221.

Yang, L., Xu, H., & Jin, Z. (2019). Estimating ground-level PM2. 5 over a coastal region of China using satellite AOD and a combined model. Journal of Cleaner Production, 227, 472-482.

Yao, F., Wu, J., Li, W., and Peng, J. (2019). A spatially structured adaptive two-stage model for retrieving ground-level PM2.5 concentrations from VIIRS AOD in China, ISPRS Journal of Photogrammetry and Remote Sensing., 151, 263-276.

Zhang, T., Zang, L., Wan, Y., Wang, W., & Zhang, Y. (2019). Ground-level PM2. 5 estimation over urban agglomerations in China with high spatiotemporal resolution based on Himawari-8. Science of the total environment, 676, 535-544.

Q2.7: Follow point 3, the authors could try to use this method on other regions where have an extensive ground-based measurements network, for example, North America, to test their validity.

Response: Thank the referee for his/her comment. At present, the study area of our study focuses on China. In the future, we will use this method on other regions, such as North America, to test its validity.

Technical corrections:

Q2.8: Section index is wrong. For example, two "3.2" sections exist in the main text.

Response: Thank the referee for his/her comment. This issue has been fixed in the manuscript.

Q2.9: Line40: should be "van Donkelaar" instead of "Van Donkelaar".

Response: Thank the referee for his/her comment. This name has been revised in the manuscript.

The main revision is as follows:

Hence, the approaches based on Chemical Transport Models (CTMs) (van Donkelaar et al., 2010; Wang et al., 2016; Weagle et al., 2018) or remote sensing satellites (Chen et al., 2018; Li et al., 2020; Stafoggia et al., 2019; Shtein et al., 2020; Wei et al., 2019; Yao et al., 2019; You et al., 2015) have been exploited to enlarge the spatial coverage of the $PM_{2.5}$ and PM_{10} monitoring.

Q2.10: Define "DUCMASS" in the main text.

Response: Thank the referee for his/her comment. The full name of "DUCMASS" has been provided in the manuscript.

The main revision is as follows:

In the meantime, the rank of DUst Column MASS density (DUCMASS) rises for the estimation of PM_{10} compared to that of $PM_{2.5}$, indicating the flexibility of our approach.